Carbonyl Complex Formation of Short-Lived Ir and Re Isotopes*

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We recently reported on the in-situ carbonyl complex formation of short-lived group 6 and group 8 elements [1]. These elements are well known to form mononuclear, volatile carbonyl complexes. However, no mononuclear, binary complexes of group 7 and 9 elements are known from literature (see e.g. [2]). We studied the transport of short-lived Ir and Re isotopes in a He-CO atmosphere, which gives a hint at the formation of volatile carbonyl complexes of these elements.

In first experiments at the TRIGA reactor in Mainz, the transport of fission products of the neutron induced fission of ^{249}Cf in a CO containing gas-stream was studied. The transported isotopes were collected on a charcoal filter and measured with a γ detector. Volatile fission products as well as short-lived isotopes of the refractory elements Mo, Tc, Ru and Rh were identified. However, precursor effects prevent a clear assignment to the transported element. To get a better understanding, the homologs Re and Ir were produced in ^{24}Mg-induced fusion reactions with ^{nat}Eu and ^{nat}Tb targets at the gas-filled separator TASCA. The recoiling isotopes were separated from the primary beam and transfer products in TASCA and thermalized in the recoil transfer chamber in a He-CO atmosphere. The volatile species were transported in the gas stream through a 10-m long capillary to a charcoal filter which was monitored by a γ detector. This way the transport of $^{178-179}$Ir and $^{170-172/172m}$Re was observed. Both, Re and Ir, apparently form volatile complexes with CO under these experimental conditions.

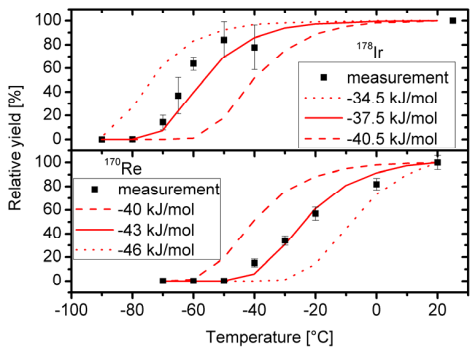

Figure 1: Transport yield of Re and Ir depending on the temperature of the isothermal chromatography column. The lines show results of Monte Carlo Simulations.

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In further investigations the adsorption and decomposition of these complexes were studied. In the adsorption-studies on a quartz surface, the gas-stream was guided through an isothermal quartz column. The temperature was varied from measurement to measurement and the transport yield through the column was determined for each temperature. These measurements were modeled with Monte Carlo Simulations, which yield values for the adsorption enthalpy of these complexes on the quartz surface. For the Re complexes the adsorption enthalpy was determined to be (−43 ± 3) kJ/mol, and for the Ir ones (−37.5 ± 3.0) kJ/mol. Figure 1 shows the breakthrough curves and the results of the Monte Carlo Simulations. The thermal stabilities of these compounds were studied by passing the gas stream through a quartz wool plug heated by an oven before reaching the charcoal filter. The transport yields were determined for various temperatures. (see Figure 2) The complexes start to decompose at temperatures around 300°C.

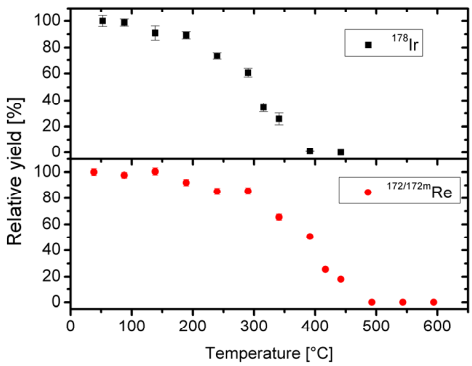

Figure 2: Transport yield of Re and Ir vs. the temperature of the quartz wool plug in the decomposition setup.

The experimental observation of the transport of Re and Ir in a CO-containing gas stream gives a hint on the formation of volatile complexes in CO. Further studies are planned to elucidate the nature of these complexes, which might open the door for bohrium and meitnerium chemistry experiments.

References

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